

PENDING CLAIMS

1. (Previously Presented) A proton-conducting polymer membrane which comprises polyazole blend and is obtained by a process (1) or (2) wherein process (1) comprises the steps

- A1) preparation of a mixture comprising polyphosphoric acid, at least one polyazole (polymer A) and/or one or more compounds which are suitable for forming polyazoles under the action of heat according to step B1),
- B1) heating of the mixture obtained according to step A1) under inert gas to temperatures of up to 400°C,
- C1) application of a layer using the mixture step B1) to a support to form a sheet-like structure,
- D1) treating said sheet-like structure formed in step C1) with hydrolysis until it is self-supporting,

or wherein process (2) comprises the steps

- A2) preparing a mixture comprising polyphosphoric acid, at least one polyazole (polymer A) and/or one or more compounds which are suitable for forming polyazoles under the action of heat according to step C2),
- B2) applying a layer using the mixture from step A2) to a support to form a sheet-like structure,

C2) heating of the sheet-like material obtained according to step B2) under inert gas to temperatures of up to 400°C,

D2) treating the sheet-like structure in step C2) with hydrolysis until it is self-supporting,

wherein at least one further polymer (polymer B) which is not a polyazole is added to the composition obtained according to step A1) or A2) and the weight ratio of polyazole to polymer B is in the range from 0.1 to 50.

2. (Previously presented) The membrane as claimed in claim 1, wherein the mixture prepared in step A1 or step A2) comprises compounds which are suitable for forming polyazoles under the action of heat in step B1) or step C2), with these compounds comprising one or more aromatic and/or heteroaromatic tetraamino compounds and one or more aromatic and/or heteroaromatic carboxylic acids or derivatives thereof which have at least two acid groups per carboxylic acid monomer and/or one or more aromatic and/or heteroaromatic diaminocarboxylic acids.

3. (Previously presented) The membrane as claimed in claim 1, wherein the mixture prepared in step A1 or step A2) comprises compounds which are suitable for forming polyazoles under the action of heat in step B1) or step C2), with these compounds being obtainable by reaction of one or more aromatic and/or heteroaromatic tetraamino compounds with one or more aromatic and/or heteroaromatic carboxylic acids or derivatives thereof which have at least two acid groups per carboxylic acid monomer or of one or more aromatic and/or heteroaromatic diaminocarboxylic acids in the melt at temperatures of up to 400°C.

4. (Previously presented) The membrane as claimed in claim 2, characterized in that aromatic and/or heteroaromatic tetraamino compounds used as compounds suitable for forming polyazoles comprise compounds selected from the group consisting of 3,3',4,4'-tetraaminobiphenyl, 2,3,5,6-tetraaminopyridine and 1,2,4,5-tetraaminobenzene.

5. (Previously presented) The membrane as claimed in claim 2, characterized in that aromatic and/or heteroaromatic carboxylic acids or derivatives thereof having at least two acid groups per carboxylic acid monomer used as compounds suitable for forming polyazoles comprise compounds selected from the group consisting of isophthalic acid, terephthalic acid, phthalic acid, 5-hydroxyisophthalic acid, 4-hydroxyisophthalic acid, 2-hydroxyterephthalic acid, 5-aminoisophthalic acid, 5-N,N-dimethylaminoisophthalic acid, 5-N,N-diethylaminoisophthalic acid, 2,5-dihydroxyterephthalic acid, 2,5-dihydroxyisophthalic acid, 2,3 dihydroxyisophthalic acid, 2,3-dihydroxyphthalic acid, 2,4-dihydroxyphthalic acid, 3,4-dihydroxyphthalic acid, 3-fluorophthalic acid, 5-fluoroisophthalic acid, 2-fluoroterephthalic acid, tetrafluorophthalic acid, tetrafluoroisophthalic acid, tetrafluoroterephthalic acid, 1,4-naphthalenedicarboxylic acid, 1,5-naphthalenedicarboxylic acid, 2,6-naphthalenedicarboxylic acid, 2,7-naphthalenedicarboxylic acid, diphenic acid, 1,8-dihydroxynaphthalene-3,6-dicarboxylic acid, bis(4-carboxyphenyl) ether, benzophenone-4,4'-dicarboxylic acid, bis(4-dicarboxyphenyl) sulfone, biphenyl-4,4'-dicarboxylic acid, 4-trifluoromethylphthalic acid, 2,2-bis(4-carboxyphenyl) hexafluoropropane, 4,4'-stilbenedicarboxylic acid, 4-carboxycinnamic acid, their C1-C20-alkyl esters, their C5-C12-aryl esters, their acid anhydrides and their acid chlorides.

6. (Previously presented) The membrane as claimed in claim 2 characterized in that the compounds suitable for forming polyazoles comprise aromatic tricarboxylic acids, their C1-C20-alkyl esters or C5-C12-aryl esters or their acid anhydrides or their acid halides or tetracarboxylic acids, their C1-C20-alkyl esters or C5-C12-aryl esters or their acid anhydrides or their acid halides.

7. (Previously presented) The membrane as claimed in claim 6, characterized in that the aromatic tricarboxylic acids comprise compounds selected from the group consisting of 1,3,5-benzenetricarboxylic acid (trimesic acid); 2,4,5-benzenetricarboxylic acid (trimellitic acid); 2-carboxyphenyl) iminodiacetic acid, 3,5,3'-biphenyltricarboxylic acid; 3,5,4'-biphenyltricarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzene-1,2,4,5-tetracarboxylic acid; naphthalene-1,4,5,8-tetracarboxylic acid, 3,5,3',5'-biphenyltetracarboxylic acid, benzophenonetetracarboxylic acid, 3,3',4,4' biphenyltetracarboxylic acid, 2,2',3,3'-biphenyltetracarboxylic acid, 1,2,5,6-naphthalenetetracarboxylic acid and 1,4,5,8-naphthalenetetracarboxylic acid.

8. (Previously presented) The membrane as claimed in claim 6, characterized in that the content of tricarboxylic acid and/or tetracarboxylic acids is from 0 to 30 mol based on dicarboxylic acid used.
9. (Previously presented) The membrane as claimed in claim 2, characterized in that the compounds suitable for forming polyazoles comprise heteroaromatic dicarboxylic acids, tricarboxylic acids and/or tetracarboxylic acids which contain at least one nitrogen, oxygen, sulfur or phosphorus atom in the aromatics.
10. (Previously presented) The membrane as claimed in claim 9, characterized in that pyridine-2,5-dicarboxylic acid, pyridine-3,5-dicarboxylic acid, pyridine-2,6-dicarboxylic acid, pyridine-2,4-dicarboxylic acid, 4-phenyl-2,5-pyridinedicarboxylic acid, 3,5-pyrazoledicarboxylic acid, 2,6-pyrimidinedicarboxylic acid, 2,5-pyrazinedicarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzimidazole-5,6-dicarboxylic acid, and also their C1-C20-alkyl esters or C5-C12-aryl esters, or their acid anhydrides or their acid chlorides are used.
11. (Previously presented) The membrane as claimed in claim 2, characterized in that the compounds suitable for forming polyazoles comprise diaminobenzoic acid and/or its monohydrochloride and dihydrochloride derivatives.
12. (Previously presented) The membrane as claimed in claim 1, wherein the polymer B) is used in step A1) or A2) in an amount in the range from 10 to 50% by weight, based on the weight of the mixture A) and/or B).
13. (Previously presented) The membrane as claimed in claim 1, characterized in that the polymer B) comprises at least one polyolefin.
14. (Previously presented) The membrane as claimed in claim 1, characterized in that the polymer B) comprises at least one polymer having C-O bonds.

15. (Previously presented) The membrane as claimed in claim 1, characterized in that the polymer B) comprises at least one polymer having C-S bonds.
16. (Previously presented) The membrane as claimed in claim 1, characterized in that the polymer B) comprises at least one polymer having C-N bonds.
17. (Previously presented) The membrane as claimed in claim 1, characterized in that the polymer B) comprises at least one inorganic polymer.
18. (Previously presented) The membrane as claimed in claim 1, characterized in that the polymer B) comprises at least one sulfonated polymer.
19. (Previously presented) The membrane as claimed in claim 1, characterized in that the heating according to step C2) is carried out after the formation of a sheet-like structure according to step B2).
20. (Previously presented) The membrane as claimed in claim 1, characterized in that the treatment according to step D1) or step D2) is carried out at temperatures in the range from 0°C to 150°C in the presence of moisture.
21. (Previously presented) The membrane as claimed in claim 1, characterized in that the treatment of the membrane in step D1) or step D2) is carried out for from 10 seconds to 300 hours.
22. (Previously presented) The membrane as claimed in claim 1, characterized in that the membrane formed after step D1) or step D2) is crosslinked by action of oxygen.
23. (Previously presented) The membrane as claimed in claim 1, characterized in that a layer having a thickness of from 20 to 4000 μm is produced in step C1) or step B2).
24. (Previously presented) The membrane as claimed in claim 1, characterized in that the membrane formed after step D1) or step D2) has a thickness of from 15 to 3000 μm .

25. (Withdrawn) An electrode provided with a proton-conducting polymer coating which comprises polyazole blends and is obtained by a process (1) or (2) wherein process (1) comprises the steps

- A1) preparation of a mixture comprising
polyphosphoric acid,
at least one polyazole (polymer A) and/or one or more compounds
which are suitable for forming polyazoles under the action of heat
according to step B1),
- B1) heating of the mixture obtained according to step A1) under inert gas to
temperatures of up to 400°C,
- C1) applying a layer using the mixture from step B1) to an electrode,
- D1) treating the electrode with the layer formed in step C1) with
hydrolysis,

or wherein process (2) comprises the steps

- A2) preparing a mixture comprising
polyphosphoric acid,
at least one polyazole (polymer A) and/or one or more compounds which
are suitable for forming polyazoles under the action of heat according to
step C2),
- B2) applying a layer using the mixture from step A2) to an electrode,
- C2) heating of the electrode with the layer obtained according to step B2)
under inert gas to temperatures of up to 400°C,

D2) treating the electrode with the layer in step C2) with hydrolysis,

wherein at least one further polymer (polymer B) which is not a polyazole is added to the composition obtained according to step A1) or A2) and the weight ratio of polyazole to polymer B is in the range from 0.1 to 50.

26. (Withdrawn) The electrode as claimed in claim 25, wherein the coating has a thickness of from 2 to 3000 μm .

27. (Withdrawn) A membrane-electrode unit comprising at least one electrode and at least one membrane as claimed in claim 1.

28. (Withdrawn) A membrane-electrode unit comprising at least one membrane as claimed in claim 1.

29. (Withdrawn) A fuel cell comprising one or more membrane-electrode units as claimed in claim 27.

30. (Withdrawn) A membrane-electrode unit comprising at least one electrode as claimed in claim 25.

31. (Withdrawn) A membrane-electrode unit comprising at least one electrode as claimed in claim 26.

32. (Previously presented) A proton-conducting polymer membrane which comprises polyazole blends and is obtained by a process (1) or (2) wherein the process (1) comprising the steps

A1) preparing a mixture comprising
polyphosphoric acid,
at least one polyazole (polymer A) and/or one or more compounds which
are suitable for forming polyazoles under the action of heat according to
step B1),

- B1) heating of the mixture obtained according to step A1) under inert gas to temperatures of up to 400°C,
- C1) applying a layer using the mixture from step A1) and/or B1) to a support to form a sheet-like structure,
- D1) treating the sheet-like structure formed in step C1) with hydrolysis until it is self-supporting,

or wherein process (2) comprises the steps

- A2) preparing a mixture comprising
 - polyphosphoric acid,
 - at least one polyazole (polymer A) and/or one or more compounds which are suitable for forming polyazoles under the action of heat according to step C2),
- B2) applying a layer using the mixture from step A2) to a support to form a sheet-like structure,
- C2) heating of the sheet-like material obtained according to step B2) under inert gas to temperatures of up to 400°C,
- D2) treating the sheet-like structure formed in step C2) with hydrolysis until it is self-supporting,

wherein at least one further polymer (polymer B) and wherein said polymer B comprises

- a) at least one polyolefin,
- b) at least one polymer having C-O bonds,

- c) at least one polymer having C-S bonds,
- d) at least one polymer having C-N bonds,
- e) at least one inorganic polymer or
- f) at least one sulfonated polymer and

said polymer B is added to the composition obtained according to step A1) or step A2) and/or step B1) and the weight ratio of polyazole to polymer B is in the range from 0.1 to 50.

33. (Previously presented) The membrane as claimed in claim 32, wherein the hydrolysis strengthens the membrane and is carried out in a temperature from 0 to 150 °C at atmospheric or superatmospheric pressure and the membrane has a thickness of 15 to 3,000 µm and the hydrolysis is carried out until the membrane has a hardness of at least 1 mN/mm².

34. (Previously presented) The membrane as claimed in claim 32, wherein the hydrolysis is carried out from 20 to 90° C in the presence of water and/or water vapor and the membrane has a thickness of 20 to 1,500 µm and the hydrolysis is carried out until the membrane has a hardness of at least 50 mN/mm².